

Infrared conductivity of photocarriers in organic molecular crystals

Chris Weber^{*1,2}, Ch. Kloc³, Michael Martin⁴, J.H. Schoen³, B. Batlogg³, Joe Orenstein^{1,2}

¹ Department of Physics, University of California, Berkeley, CA 94720

² Material Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720

³ Bell Laboratories, Lucent Technologies, Murray Hill, New Jersey 07974-0636

⁴ Advanced Light Source Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720

This project, begun in March of 2001, seeks use infrared spectroscopy to probe the properties of photocarriers in organic molecular crystals. A group at Bell Laboratories has recently succeeded in creating FETs on ultrapure single crystals of pentacene, tetracene, and other “polyacenes” (rigid, rodlike chains of Benzene rings). Carriers injected into these transistors have exhibited metal-insulator transitions, superconductivity, lasing, and the quantum Hall effect (see, e.g., J. H. Schoen, Ch. Kloc, B. Batlogg, *Science* v. 288 p. 2338; *Science* v. 288 p. 656; *Nature* v. 406 p. 702). The goal of our experiment is to photoexcite carriers in these same materials, and to measure the infrared spectrum of these photocarriers.

The apparatus, at ALS beamline 1.4, includes an Argon-ion laser for photoexcitation of charge carriers, a Bruker 66v/S Fourier-Transform Infrared (FTIR) spectrometer, and a variable-temperature cryostat. The laser allows photoexcitation at energies from 2.4 eV to 3.5 eV. At the UV frequencies, the quantum efficiency of photocarrier generation in pentacene is high, about 30%. The light is coupled to the sample (in the cryostat) by means of an optical fiber. The cryostat allows us to reach temperatures from 5 K to 300 K, and the FTIR spectrometer allows measurement of infrared transmission on the range of at least 100 cm^{-1} to 7800 cm^{-1} .

The basic result of a measurement is a transmission spectrum, $T(\omega)$, either with the sample illuminated with laser light, $T_{\text{on}}(\omega)$, or with the sample unilluminated, $T_{\text{off}}(\omega)$. From these spectra we determine difference spectra, $\Delta T / T = (T_{\text{on}}(\omega) - T_{\text{off}}(\omega)) / T_{\text{off}}(\omega)$. Our measurements at room temperature on crystals of Tetracene have shown good reproducibility, both in the shape and in the magnitude of the difference spectra. The measurement is sensitive to changes in the transmission of one part in 10^4 or better through most of the spectral range. We have resolved many clear and reproducible features that are an order of magnitude larger than the noise.

Although these results demonstrate that the apparatus is, indeed, able to make the desired measurements, they do not put us particularly close to our scientific goals. Most or all of the features we have seen thus far appear to be due to shifting or broadening of phonon absorptions due to the heating effect of the laser. This result at room temperature is no surprise, as the mobility of carriers in polyacene crystals increases as T^{-2} below room temperature, so that the conduction of the carriers should only become visible at low temperatures. More surprising was the result that tetracene crystals large enough to measure optically will invariably shatter upon cooling below about 180 K (the temperature seems to vary a bit). Pentacene crystals, on the other hand, do not shatter, but also do not seem to be available in sizes large enough for optical measurement. Other

polyacenes have much lower efficiencies of photocarrier generation (due to their having broader bandgaps), and so are not suited to this experiment.

We are now seeking thin-film samples of the polyacenes, as these should survive cooling and should have the large area desirable for infrared measurement. High-quality pentacene films have been shown to display many of the same electronic properties as their single-crystal counterparts (J. H. Schoen, Ch. Kloc, *Applied Physics Letters*, v. 79, p. 4043).

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* To whom correspondence should be addressed:
cpweber@lbl.gov
(510) 486-5879